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# Ion behavior and gas mixing in electron cyclotron resonance plasmas as sources of highly charged ions

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This article deals with ion confinement in small open-ended magnetic devices, the electron cyclotron resonance ion sources (ECRIS) that were developed for multicharged ion production. The ECRIS are basically ECR-heated plasma confinement machines with hot electrons and cold ions. The main parameters of the ion population in ECRIS plasmas are successively analyzed, temperature, collisions, losses, ionization, confinement times, charge state distribution equilibrium, followed by the analysis of the gas mixing effect, a specific technique to improve the performance as an ion source. A series of experiments is described for the systematic analysis of the phenomena related to gas mixing. It is shown that high charge state optimization by gas mixing relies on a compromise between three criteria, ion losses, mass effect, and ionization rates. The article stresses the role of some fundamental plasma parameters for the next generation of high charge state/high intensity ion sources.

## I. INTRODUCTION

Electron cyclotron resonance ion sources (ECRIS) are small mirror machines [?] equipped with a radial minimum-B magnetic multipole, usually hexapolar and made of permanent magnets (Fig. ??). The electrons are heated by interacting with rf waves at cyclotron resonance on one of the closed equal- $|B|$  surfaces of such a magnetic configuration, the equal- $|B|_{ecr}$  surface defined by

$$\omega_{ce} = \frac{e |B|_{ecr}}{m_e} = \omega_{rf} \quad .$$

The extracted ion currents at one end of the device are actually the ion losses of the trapped plasma. Several comprehensive references about ECRIS have been recently published [?,?] ; the theoretical and experimental studies of electrons in an ECRIS plasma will be found in [?]. The present article puts more emphasis on ion parameters.

Basically an ECRIS has the property of confining a hot electron plasma, its main components are: (i) the magnetic configuration, i.e. the plasma container, (ii) the rf power input, i.e. the source of energy that heats up and sustains the plasma electrons in the magnetic trap, (iii) the internal (ionization) and external sources of electrons, which allow the electron density to build up, (iv) the injected neutrals (gas or metal), i.e. the fuel injected in order to compensate for the ion losses and to control the neutral pressure. The ion current  $I_i^q$  of species  $i$  and charge state  $q$ , extracted at one end of an ECRIS, is equal to the ion loss rate

$$I_i^q \simeq \frac{1}{2} \frac{n_i^q q e V_{ex}}{\tau_i^q} \quad , \tag{1}$$

here  $n_i^q$  is the density of ions of species  $i$  and charge state  $q$ ,  $\tau_i^q$  the ion confinement time ;  $V_{ex}$  is the part of the hot plasma volume that maps along the magnetic field lines into the extraction area. Assuming plasma charge neutrality, one may expect that, approximately,

$$I_i^q \propto n_e V_{ex} \quad .$$

The total electron density  $n_e$  consists of thermal electrons  $n_{eth}$ , and hot electrons  $n_{eh}$ , such that  $n_e = n_{eth} + n_{eh}$ , with  $n_{eh} \ll n_{eth}$  at low rf power and  $n_{eh} \gg n_{eth}$  at high rf power. These two populations may be abusively described

by two different maxwellian distributions of temperatures  $T_{eth}$  and  $T_{eh}$ . For the sake of simplicity a single electron distribution of temperature  $T_e$  will be considered in this article.

Optimizing an ECRIS in order to obtain high charge state/high intensity ion currents, may look like a contradictory task : how to maintain both good confinement (large confinement times  $\tau_i^q$  for high charge states) and high losses (high intensity extracted ion currents  $I_i^q$ )? This contradiction is also included in formula (??). However because of plasma confinement self-consistency, the dependence of particle density with particle confinement time is not linear, and usually increasing the confinement time increases both particle density and loss rate. Owing to the large number of parameters involved in ECRIS operation, i.e. the various ECRIS knobs, rf power, gas pressure, magnetic field, etc..., it is possible to find a compromise, the well-known tuning of ECRIS. Note that this tuning is always necessary in ECRIS, and is different for every ion. When running an ECRIS the operator plays to some extent with these different knobs in order to improve the desired extracted ion intensity.

The present article aims to analyze the particular aspects of ions in these ECR plasmas.

## II. MAIN ION PARAMETERS IN ECRIS

The main ion parameters of ECR multi-species plasmas, such as temperature, collision frequencies, confinement time, are evaluated in this section.

The non-self-consistent fluid description of ions in ECRIS involves three dominant processes, step-by-step ionization by electron impact, charge exchange, and diffusion or transport (note that this last term is the important term for the users of an ion source). Ions are considered to be maxwellian owing to their high collisionality (see below). The evolution in time of the ion density  $n_i^q$  is given by

$$\begin{aligned} \frac{dn_i^q}{dt} = & +n_e \langle \sigma v \rangle_{q-1 \rightarrow q}^{ion} n_i^{q-1} - n_e \langle \sigma v \rangle_{q \rightarrow q+1}^{ion} n_i^q \\ & + n_{0_i} \langle \sigma v \rangle_{q+1 \rightarrow q}^{cx} n_i^{q+1} - n_{0_i} \langle \sigma v \rangle_{q \rightarrow q-1}^{cx} n_i^q \\ & - \frac{n_i^q}{\tau_i^q} , \end{aligned} \quad (2)$$

here  $n_{0_i}$  is the neutral density of atoms of species  $i$ . The ion temperature  $T_i^q$  of ions of species  $i$  and charge  $q$  is given by the ion energy balance equation

$$\frac{d(n_i^q T_i^q)}{dt} = n_e (T_e - T_i^q) \nu_{eq} \rightarrow q - \frac{n_i^q T_i^q}{\tau_{T_i^q}} . \quad (3)$$

In the righthand side of this equation, the first term (electron-ion energy equipartition rate) is the energy taken by the ions ( $i, q$ ) from the electron population, note that  $T_e \gg T_i^q$ ; the second term stands for the ion energy diffusion (or transport) loss rate,  $\tau_{T_i^q}$  being the ion energy confinement time. The ion-ion collision frequency  $\nu_{ij}$  is so high that all ions have actually the same temperature  $T_i = T_i^q$ . Therefore all equations (??) may be summed up to give in steady state ( $\frac{d}{dt} = 0$ )

$$n_e T_e \nu_{eq} \rightarrow i = T_i \sum_i \sum_q \frac{n_i^q}{\tau_{T_i^q}} \approx T_i \sum_i \sum_q \frac{n_i^q}{\tau_i^q} , \quad (4)$$

where we use  $\tau_i^q$ , the ion confinement time, as an approximation of the ion energy confinement time  $\tau_{T_i^q}$ . The electron-ion energy equipartition frequency,  $\nu_{eq} \rightarrow i$ , is given by the following formula [?]

$$\nu_{eq} \rightarrow i \simeq \frac{3.2 \cdot 10^{-9} \ln \Lambda_{ei}}{T_e^{3/2}} \sum_i \frac{\sum_q n_i^q q^2}{A_i} \quad (\text{s}^{-1}, \text{eV}, \text{cm}^{-3}), \quad (5)$$

where  $A_i$  is species  $i$  mass number. Owing to the large ratio of ion to electron masses, this electron-ion collisional heating is not very efficient :  $T_i$  is usually of the order of 1 eV as measured by Doppler broadening of HeII line [?]. The ion-ion collision frequency  $\nu_{ij}$  is given by the expression [?](using the same units,  $\text{s}^{-1}$ , eV,  $\text{cm}^{-3}$ )

$$\nu_{ij} = \frac{1}{\tau_{ij}} \simeq \frac{6.8 \cdot 10^{-8} \ln \Lambda_{ij} q^2}{T_i^{3/2} A_i} \sum_j \sqrt{A_j} \sum_q n_j^q q^2 . \quad (6)$$

Orders of magnitude of these ion parameters for charge  $q=10$  are given in table ?? : we consider a typical pure argon plasma in a 14 GHz ECRIS ( $B_{min} \simeq 0.4$  T) with  $T_i=1$  eV,  $T_e=500$  eV and  $n_e=5.10^{11}\text{cm}^{-3}$  according to experimental data [?], mean plasma effective ion charge  $q_{eff}=8$  ( $=\sum_q n_i^q q^2/n_e$ ), assuming both  $\ln \Lambda_{ei}$  and  $\ln \Lambda_{ii} \simeq 10$  [?] ;  $f_{ci}$  is the ion cyclotron frequency, and  $\rho_i$  the ion gyroradius. Here  $\Delta\ell=v_{T_i}/\nu_{ii}$  is the average distance between two collisions along the ion trajectory,  $v_{T_i}=\sqrt{kT_i/m_i}$  being the ion thermal velocity.

The collision rate and the temperature of highly charged ions are important parameters for their confinement, and therefore their production. The ion confinement time may be evaluated by three different methods.

(i) In most cases as shown in table ?? the high ion collisionality makes ions almost *unmagnetized* : as  $\Delta\ell/2\pi\rho_i \ll 1$ , ions experience a random walk diffusion. In this collisional regime the ion confinement time  $\tau_i^q$  may be calculated as follows : during  $\tau_i^q$  the ion moves over the characteristic plasma dimension  $a$ , while undergoing  $N$  ( $=\tau_i^q/\tau_{ij}$ ) collisions, therefore statistically

$$a \simeq \sqrt{N} \Delta\ell ,$$

which gives the following formula for  $\tau_i^q$  in the units (s, cm, eV,  $\text{cm}^{-3}$ )

$$\tau_i^q \simeq 7.1 \cdot 10^{-20} a^2 \ln \Lambda_{ij} \frac{q^2}{T_i^{5/2}} \sum_j \sqrt{A_j} \sum_q n_j^q q^2 . \quad (7)$$

In today ECRIS the dimension  $a$  ( $\sim$  plasma length, ECR surface diameter) is only a few centimeters long ( $\leq 5$  cm), for  $\text{Ar}^{10+}$  ions as an example  $\tau_i^q$  is typically of the order of 10 ms.

(ii) Experimental measurements of ion densities from VUV spectroscopy [?] in ECR plasmas may suggest a linear ion charge dependence of ion confinement time. If the ion motion is governed by mobility rather than diffusion [?], a linear ion charge dependence of  $\tau_i^q$  is actually found as  $\tau_i^q$  may be calculated as follows

$$\tau_i^q \simeq \frac{a}{\mu_i E_\phi} ,$$

here  $\mu_i$  is the ion mobility, and  $E_\phi$  is the electric field within the plasma that results from the plasma potential  $\phi$  [?]

$$\mu_i = \frac{qe}{m_i \nu_{ij}} , \quad E_\phi = -\nabla\phi .$$

Using formula (??) for  $\nu_{ij}$  yields  $\tau_i^q$  of the form

$$\tau_i^q \simeq k_\mu a \frac{q}{T_i^{3/2}} \sum_j \sqrt{A_j} \sum_q n_j^q q^2 , \quad (8)$$

where the coefficient  $k_\mu$  is proportional to  $(\nabla\phi)^{-1}$ , note also that  $\phi$  depends upon the input rf power. Actually it is likely that the use of formulas (??) or (??) depends on whether the average effect of electrical forces dominates over that of diffusion forces along the ion trajectory or inversely. Different plasma regions, e.g. central plasma and sheath, or different plasma regimes would lead to using (??) and/or (??) accordingly.

(iii) Other formulas proposed for  $\tau_i^q$  are derived from calculations made for mirror trapped ions in a potential well [?, ?, ?]. High energy electrons magnetically confined in ECRIS could create a small negative potential dip  $\Delta\phi$  at the center of the plasma where the plasma potential  $\phi$  is positive [?, ?]. For low-Z ions, when collisions are less frequent ( $\nu_{ij} \sim$  bounce frequency of ions between mirrors), this negative dip of potential  $\Delta\phi$  leads to the flow rate formula for  $\tau_i^q$

$$\tau_i^q \simeq \tau_f = R \frac{a}{v_{T_i}} \exp\left(\frac{|qe \Delta\phi|}{kT_i}\right) , \quad (9)$$

$R$  being the mirror ratio ( $=B_{max}/B_{min}$ ). Different methods have been proposed to evaluate the ratio  $\Delta\phi/T_i$  [?, ?, ?, ?], which is found  $\ll 1$ ,  $\Delta\phi$  being much lower than 1 volt.

### III. ION CHARGE STATE DISTRIBUTION

In the right handside of equation (??), the last term, diffusion or transport, is the dominant loss process. For low and medium charge states,  $q \leq q_M$ ,  $q_M$  being the peak of the charge distribution of extracted ion currents ( $\simeq 10$  for

argon in today ECRIS), charge exchange may be neglected as a loss process with respect to transport. Therefore in steady state ( $\frac{d}{dt} = 0$ ), equation (??) shows that the ion charge state distribution approximately results from the balance between ionization and transport

$$n_e < \sigma v >_{q-1 \rightarrow q}^{ion} n_i^{q-1} - n_e < \sigma v >_{q \rightarrow q+1}^{ion} n_i^q \simeq \frac{n_i^q}{\tau_i^q} ,$$

from which it is possible to obtain the derivative of the ion current  $(\Delta I)_i^q = I_i^q - I_i^{q-1}$  as a function of charge state  $q$

$$\frac{(\Delta I)_i^q}{I_i^{q-1}} \approx \frac{n_i^q}{n_i^{q-1}} - 1 \simeq \frac{\tau_i^q - \tau_{ion}^q}{\tau_{ion}^q (1 + \tau_i^q n_e < \sigma v >_{q \rightarrow q+1}^{ion})} ,$$

where  $\tau_{ion}^q = (n_e < \sigma v >_{q-1 \rightarrow q}^{ion} - n_e < \sigma v >_{q \rightarrow q+1}^{ion})^{-1}$  is the ionization time for charge state  $q$ . Therefore the peak ( $q_M$ ) of the charge state distribution (CSD) of the extracted ion currents, i.e. the most abundant extracted current, may be approximately interpreted as the crossover of the ion confinement time and the ionization time (which varies according to the atomic shells M,L,K) as functions of charge state,  $\tau_i^{q_M} \simeq \tau_{ion}^{q_M}$  (Fig. ??).

Figure ?? shows that an increase of  $q_M$ , and therefore of source performance, may be obtained either (i) by decreasing  $\tau_{ion}^q$ , which means increasing  $n_e$ ; or (ii) by increasing  $\tau_i^q$ , which means decreasing  $T_i$  and increasing  $n_e$  according to formulas (??-??). The effect of charge exchange losses, which can be integrated into  $\tau_{ion}^q$ , is also shown in figure ??.

Figure ?? illustrates the ion current CSD evolution from Caprice source (a) to SERSE source (b). Caprice [?] is a compact high performance source ( $a \simeq 4$  cm,  $B_{max} \simeq 1.5$  T), its ion current CSD peaks on charge 10/11. SERSE [?] is a high magnetic field superconducting source ( $a \simeq 6$  cm,  $B_{max} \simeq 2.7$  T), one of the most performant today ECRIS, that has both a high electron density because of its magnetic field and large confinement times because of its dimensions. As expected from formulas (??-??) SERSE ion current CSD peaks on charge 12/13.

#### IV. GAS MIXING EFFECT IN ECRIS

This remarkable effect, independently discovered by two ECRIS pioneers [?,?], consists of mixing a lighter gas to the main element of the ECR discharge, in order to increase the output currents of highly charged ions of this element. The density of the gas being added is usually important with respect to that of the main element, and its mass is always lower. Many efforts and works [?,?, ?, ?, ?, ?, ?] concentrated on the understanding of this effect which was given several interpretations, none of them being fully satisfactory. (i) A dilution effect lowering the mean ion charge [?], this would reduce the electron loss rate, improve the electron density and the source performance. (ii) Ion cooling resulting from the mass effect in ion-ion collisions, the low mass/low charge ions of the added gas that drag energy from heavy ions, would efficiently carry out of the plasma the ion energy because of their lower confinement [?,?]. (iii) An increase of the electron density because of the better ionization efficiency of the added gas, the conditions  $n_e$ ,  $T_e$  of the ECRIS plasma with added gas being more favorable to the desired multicharged ion production [?]. (iv) An increase of the plasma stability [?,?] : low frequency (a few Hz) relaxations [?], the origin of which could be electron cooling caused by sputtered wall metal atoms from plasma ions accelerated by potential  $\phi$  [?], would reduce the electron/ion confinement; these relaxations would get stabilized by decreasing  $\phi$ . Actually as a result of gas mixing, the plasma potential  $\phi$  is usually observed to decrease [?] : this is related to the higher mobility of the added gas ions, the addition of light ions increases the average ion mobility of the ECR plasma.

There are many experimental evidences that a mass effect exists in the gas mixing technique [?], and ion cooling, i.e.  $T_i$  decrease which increases the ion confinement time as shown in formulas (??) and (??), is likely a major consequence of gas mixing, although any direct measurement of this cooling has not been achieved. However inversely ion heating by ion cyclotron waves has been observed to deteriorate the high charge state performance of an ECRIS plasma [?,?].

Whether or not the beneficial effect of gas mixing would result in decrease of  $T_i$ , a series of experiments on the Caprice source [?] discussed below was carried out.

##### A. Model for processing data

The method employed to process the data makes use of the formulas given in section ??. We shall make the assumption that  $\tau_i^q$  is given by formula (??). Using formula (??) and formula (??) which is rewritten as

$$\tau_i^q \simeq k_c a^2 \frac{q^2}{T_i^{5/2}} \sum_i \sqrt{A_i} \sum_q n_i^q q^2, \quad (10)$$

where  $k_c = 7.1 \cdot 10^{-20} \ln \Lambda_{ij}$  is a constant, formula (??) becomes

$$T_i^{7/2} = k_{ei} k_c a^2 \left( \frac{n_e}{\sqrt{T_e}} \right) \left( \sum_i \sqrt{A_i} \sum_q n_i^q q^2 \right) \left( \frac{\sum_i \frac{1}{A_i} \sum_q n_i^q q^2}{\sum_i \sum_q \frac{n_i^q}{q^2}} \right), \quad (11)$$

here  $k_{ei} = 3.2 \cdot 10^{-9} \ln \Lambda_{ei}$  is a constant. Note that all the ion species, both main element and mixing gas, as well as any other species existing in the ECR plasma, are included in formulas (??) and (??). We write  $I_i^q$  from formula (??) in the form

$$I_i^q = k_L \frac{n_i^q q}{\tau_i^q}, \quad (12)$$

$k_L$  is a constant which is not known with precision (fraction of plasma losses being extracted). The mean ion charge  $q_{eff}$  and  $T_i$  can be calculated as functions of the extracted ion currents  $I_i^q$  after some algebra

$$\begin{aligned} n_e &= \sum_i \sum_q n_i^q q = \frac{k_c a^2}{k_L} \frac{\sum_i \sqrt{A_i} \sum_q n_i^q q^2}{T_i^{5/2}} \sum_i \sum_q I_i^q q^2, \\ q_{eff} &= \frac{1}{n_e} \sum_i \sum_q n_i^q q^2 = \frac{\sum_i \sum_q I_i^q q^3}{\sum_i \sum_q I_i^q q^2}, \\ T_i^{7/2} &= k_{ei} k_c a^2 \left( \frac{n_e^2 q_{eff}}{\sqrt{T_e}} \right) \left( \frac{\sum_i \sqrt{A_i} \sum_q I_i^q q^3 \cdot \sum_i \frac{1}{A_i} \sum_q I_i^q q^3}{\sum_i \sum_q I_i^q q^3 \cdot \sum_i \sum_q \frac{I_i^q}{q}} \right). \end{aligned} \quad (13)$$

The advantage of formula (??) is its normalized form showing explicitly the expression  $\left( \frac{n_e^2 q_{eff}}{\sqrt{T_e}} \right)$  which is proportional to the absorbed rf power (by electrons), as long as there are no other electron losses than the collisional ones, particularly no rf induced electron losses (rf pitch angle scattering [?]) which may occur at high rf power and cause saturation of extracted currents. At low and medium input rf powers, this expression is close to the input rf power, provided that there is no reflected power towards the rf generator and no direct rf losses, e.g. radiated rf power from the extraction hole or rf losses in the coupling system. If these conditions are satisfied, this expression  $\left( \frac{n_e^2 q_{eff}}{\sqrt{T_e}} \right)$  is given by the electron power balance equation

$$\frac{d(n_e T_e)}{dt} = \frac{P_{abs}}{V_p} - \frac{n_e T_e}{\tau_{T_e}} - n_e (T_e - T_i) \nu_{eq \rightarrow i} - P_{ion}.$$

In the right handside of this equation, the third term which is the ion heating term used above in Eq.(??) as well as the last term, the power consumed to ionize, are both negligible as compared to the other terms. Therefore in steady state ( $\frac{d}{dt} = 0$ ), this equation reduces to the following equation,  $P_{abs}$  being the absorbed rf power and  $V_p$  the plasma volume,

$$\frac{n_e T_e}{\tau_{T_e}} = \frac{P_{abs}}{V_p}. \quad (14)$$

$\tau_{T_e}$  is the electron energy confinement time which is approximately given by the expression

$$\tau_{T_e} \simeq 5.2 \cdot 10^3 \frac{T_e^{3/2}}{n_e q_{eff}} \quad (\text{s, eV, cm}^{-3}). \quad (15)$$

Combining Eqs.(??) and (??) yields

$$\left( \frac{n_e^2 q_{eff}}{\sqrt{T_e}} \right) \simeq \frac{5.2 \cdot 10^3}{e} \frac{P_{abs}}{V_p} \quad (\text{C, W, cm}^3), \quad (16)$$

where  $e = 1.6 \cdot 10^{-19}$  C. For Caprice source data that are analyzed below,  $V_p \simeq 100 \text{ cm}^3$  and  $a \simeq 4 \text{ cm}$ .

Extracted ion currents from the Caprice source are separated by magnetic analysis, overlapping ion currents (of same  $q/m$ ) being evaluated by interpolation. While optimizing the extracted current of a desired highly charged argon ion, series of runs were achieved in different gas conditions : (i) pure argon, (ii) argon and oxygen 16, (iii) argon and oxygen 18, (iv) argon and neon 22. All source parameters were maintained constant except for the gas pressures of both argon and added gas that were adjusted for the optimization. For a few extracted Ar ion currents optimized, ( $\text{Ar}^{14+}$ ,  $\text{Ar}^{11+}$ ,  $\text{Ar}^{8+}$ ), table ?? gives experimental data, extracted Ar ion currents  $I_{Ar}^q$  (in  $\mu\text{A}$ , electrical microAmpere for charge states 8/11/14, out of which only one is optimized), total extracted particle current  $\sum_q I_i^q/q$  (total ion loss rate,  $1 \mu\text{A}=6.25 \cdot 10^{12}$  part./s), the expression  $P_{ei} = \sum_i \sum_q I_i^q q^3 / A_i$  which accounts for the ion energy from electrons, and calculated  $T_i$  from formula (??). *Each line of table ?? corresponds to a different plasma*, the three ion currents being shown give some idea of argon ion current distribution ; note that  $P_{ei}$  depends upon the ion densities of the corresponding plasma. Before going on to data analysis, a few specific experimental aspects of ECRIS plasmas are worth mentioning : electron density and temperature increase with rf power [?], gas mixing technique is observed to be efficient for increasing only high charge state ion currents [?], and not for low charge state ion currents. Obviously in the optimization process of an ion current by changing gas pressures, both  $n_e$  and  $T_e$  of the plasma vary. According to formula (??)  $T_i$  decreases with decreasing rf power.

The basic effect of gas mixing can be seen in table ?? by comparing the optimized currents  $I_{Ar}^q$  in pure argon with those obtained in different gas mixtures : gas mixing is efficient for  $\text{Ar}^{14+}$  ions, less efficient for  $\text{Ar}^{11+}$  ions and almost not efficient at all for lower charge state ions such as  $\text{Ar}^{8+}$ . The behavior of ion currents as a function of rf power may also result from changes of  $n_e$  and  $T_e$ , this last parameter controls the different charge state ionization cross-sections.

Let us consider first  $\text{Ar}^{14+}$  ion current optimization. From table ?? gas mixing induces ion cooling, which likely causes the  $\text{Ar}^{14+}$  ion density and current increases. Actually a  $T_i$  decrease is the result of two effects : lowering of  $P_{ei}$  (less energy to the ion population), and increase of  $\sum_q I_i^q/q$  (more losses of the ion population). On one hand gas mixing definitely increases the total ion loss rate : it adds low charge ions that have high losses, this is particularly efficient for high charge ion production like  $\text{Ar}^{14+}$ . On the other hand  $P_{ei}$  depends upon several parameters, ion densities (which means also  $n_e$  and  $T_e$ ) and ion masses. The ion mass effect is clear for  $\text{Ar}^{14+}$  ions while comparing together mixtures (ii), (iii), and (iv) . The ion density effect appears also in comparing mixtures (ii) or (iii) with mixture (iv), neon having lower ionization rates than oxygen (Fig.?? from calculations [?]). As a result the electron density of an argon/neon plasma is lower than that of an argon/oxygen plasma, therefore  $P_{ei}$  is lower for neon, but this reduces also the delivered  $\text{Ar}^{14+}$  ion current in spite of the low  $T_i$ . This shows that in gas mixing several effects which may counterbalance each other are mixed up. Of course in the quantitative discussion given here, wall effects are not taken into consideration.

Low charge state ions have normally larger loss rates because of lower confinement times, therefore the dominant effect of loss rate change shown in table ?? between pure argon (i) and other gas mixtures (ii), (iii) and (iv) when optimizing  $\text{Ar}^{14+}$  ion currents, does not exist when optimizing  $\text{Ar}^{8+}$  ions and ion cooling is not as evident as when optimizing  $\text{Ar}^{14+}$  ions. Therefore an argon plasma optimizing a low charge state ion does not need much added gas, and is almost a pure argon plasma ; this is shown in table ?? ( $\text{Ar}^{8+}$  ion optimization) that gives for the same plasmas as in table ??, the mean plasma ion charge  $q_{eff}$ , the mean argon ion charge  $q_{Ar} = (\sum_q I_i^q q^3 / \sum_q I_i^q q^2)_{Ar}$ , the mean added gas ion charge  $q_{ag} = (\sum_q I_i^q q^3 / \sum_q I_i^q q^2)_{ag}$ , and the mean percentages in the plasma of these two gases  $\alpha_{Ar}$ ,  $\alpha_{ag}$  calculated from these mean ion charges.

The mean percentages of the two gases were also calculated by using the linear  $q$  dependency assumption of the ion confinement time with formula (??), the expressions of  $q_{Ar}$  and  $q_{ag}$  being  $q_{Ar} = (\sum_q I_i^q q^2 / \sum_q I_i^q q)_{Ar}$ ,  $q_{ag} = (\sum_q I_i^q q^2 / \sum_q I_i^q q)_{ag}$ . The resulting added gas percentages are approximately at most a factor 2 higher than those given in table ?. Although these percentages calculated in the linear  $q$  dependency assumption sound closer to the usually estimated experimental values (from pressures), the numbers of table ?? reported here are calculated in the square  $q$  dependency assumption for consistency because some quantities of table ??, e.g.  $T_i$ , could not be calculated in the linear  $q$  dependency assumption (unknown coefficient  $k_\mu$  because no measurement of  $E_\phi$ ). Note that the linear  $q$  dependency assumption of the ion confinement time would also lead to ion cooling.

As mentioned above, it is also likely that adding another gas in a pure argon plasma, causes variations of important plasma parameters such as  $n_e$  and  $T_e$ . This might explain the opposite effect of rf power and gas addition on mixtures (ii) and (iii) in table ?? : as the rf power increases (which increases  $n_e$  and  $T_e$ ), the amount of added gas for best optimization has to be reduced.

This article has shown the importance of a few plasma parameters, ion temperature, ion collision rate, and – to some extent – plasma dimensions, in the control of ECRIS ion confinement times for high charge state ion density/loss rate increase. Specifically the systematic study of gas mixing presented above, e.g. mixing oxygen into an argon plasma, for the production of more intense high charge state ion currents (loss rates), together with earlier studies of the authors and other observations cited above, give credit to the effect of ion temperature decrease by gas mixing optimization. To summarize this analysis, several mechanisms may be accounted for in the gas mixing technique :

- the high losses of the added gas ions ; ideally the added gas, e.g. oxygen, has only low charge states that have high losses as compared to those of the main element, e.g. argon, of which high charge state losses have to be increased. This refers to enhanced ion energy losses by dilution.

- the mass effect of the added gas ; ideally the added gas mass is as close to that of the main element as possible, not to increase the ion energy from electrons, and to allow for easy exchange of energy with main element ions. This refers to minimizing the energy equipartition between electrons and ions, and to the strong coupling by collisions between all ions.

- however practising the gas mixing technique may jeopardize the plasma electron density and therefore reduce all the ion densities, because of the lower ionization rates of the added gas (Fig.??) with respect to those of the main element.

Obviously there is some contradiction – not to say impossibility – in fulfilling together these three criteria, and gas mixing always results from a compromise. Therefore it is interesting to analyze a few well known actual situations. From the above given arguments it follows that mixing xenon into an argon plasma will not help at all to produce high charge states of argon, because of violation of the first criterion ; one might expect nitrogen not as convenient as oxygen for mixing into an argon plasma, the second and third criteria being not fulfilled as well, this is experimentally observed ; hydrogen does not work for gas mixing because of violation of the second and third criteria, it behaves like an ion heater as it receives more energy from electrons than any other ion, moreover it has a low ionization rate. For usual elements, metal and gas ions, oxygen 16 as a mixing gas should be the most convenient according to the first and third criteria. These observations are in agreement with general experience [?, ?, ?]. As described above and previously observed [?] oxygen 18 is actually the best candidate for high charge state optimization by gas mixing into an argon plasma : it gives the best compromise for both ion losses, mass effect and ionization rate.

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FIG. 1. Sketch of an ECR ion source :  $|B|_{last}$  refers to the highest closed  $|B|$  surface of electron confinement.

FIG. 2. Diagram showing how to determine the charge state  $q_M$  of the most abundant ion current extracted from ECRIS, from the crossover of ionization and ion confinement times. The arrows indicate how to change these times in order to increase  $q_M$ . The effect of charge exchange losses (cx) is indicated.

FIG. 3. Argon ion current distributions delivered by (a) Caprice source, (b) SERSE source, while optimizing  $\text{Ar}^{16+}$  ion current with  $^{16}\text{O}$  as a mixing gas.

FIG. 4. First ionization rates of a few gases.

TABLE I. Typical Ar<sup>10+</sup> ion parameters.

$\nu_{ii}(\text{s}^{-1})$	$\nu_{eq\,ei}(\text{s}^{-1})$	$f_{ci}(\text{s}^{-1})$	$\rho_i(\text{mm})$	$\Delta\ell(\text{mm})$
43. 10 <sup>6</sup>	0.28	1.5 10 <sup>6</sup>	0.16	0.04

TABLE II. Data of gas mixing experiments : plasma conditions, gas mixtures (i) pure Ar, (ii) Ar + <sup>16</sup>O, (iii) Ar + <sup>18</sup>O, (iv) Ar + <sup>22</sup>Ne, and input rf power settings. Measured extracted Ar ion currents in (eμA) when optimizing only one charge state (that of bold numbers), Ar<sup>14+</sup> (top table), Ar<sup>11+</sup> (mid-table), Ar<sup>8+</sup> (bottom table). Calculated parameters : total particle loss rate  $\sum I_i^q/q$  in (pμA), expression (ion energy from electrons)  $P_{ei} = \sum_i \sum_q I_i^q q^3/A_i$ , ion temperature  $T_i$  from formula (??).

gas	P <sub>HF</sub> (W)	I <sub>Ar</sub> <sup>q</sup> (eμA) 8/11/14	$\sum I_i^q/q$ (pμA)	P <sub>ei</sub> (a.u.)	T <sub>i</sub> (eV)
(i)	800	132/31/ <b>0.68</b>	265.	8134.	2.29
	600	145/32/ <b>0.66</b>	246.	8615.	2.19
(ii)	800	43.6/47/ <b>5.6</b>	353.	8477.	2.13
	600	49/46.6/ <b>4.2</b>	366.	8846.	1.95
	400	57/23/ <b>0.60</b>	415.	7513.	1.58
	200	53/13/ <b>0.23</b>	364.	4609.	1.18
(iii)	800	23/35/ <b>7.0</b>	340.	7691.	2.07
	600	32/35/ <b>5.0</b>	335.	7929.	1.93
	400	32/15/ <b>0.80</b>	365.	4683.	1.43
(iv)	800	19/20.4/ <b>2.6</b>	320.	5736.	1.93
(i)	800	130/ <b>31</b> /0.60	264.	8116.	2.29
	600	144/ <b>33</b> /0.70	246.	8755.	2.20
	400	182/ <b>20</b> /0.15	262.	8514.	1.91
	200	110/ <b>2.4</b> /0.0	285.	4358.	1.26
(ii)	800	116/ <b>54</b> /2.2	355.	10890.	2.28
	600	76/ <b>56</b> /4.0	328.	10058.	2.10
	400	93/ <b>37</b> /1.0	361.	8817.	1.75
	200	82/ <b>15</b> /0.14	339.	5294.	1.26
(i)	800	<b>181</b> /5.2/0.0	405.	7285.	1.97
	600	<b>165</b> /2.5/0.0	414.	6797.	1.77
	400	<b>187</b> /14/0.08	267.	8015.	1.87
	200	<b>112</b> /2.5/0.0	284.	4435.	1.27
	50	<b>28</b> /0.03/0.0	212.	1606.	0.70
(ii)	800	<b>160</b> /51.3/1.1	321.	11390.	2.39
	600	<b>160</b> /38/0.7	326.	10166.	2.12
	400	<b>200</b> /20/0.1	304.	9387.	1.88
	200	<b>136</b> /5.4/0.03	319.	5399.	1.30
	50	<b>51</b> /0.0/0.0	292.	2291.	0.70
(iii)	800	<b>182</b> /6.6/0.0	498.	7130.	1.84
	600	<b>190</b> /9.3/0.0	515.	7629.	1.71
	400	<b>160</b> /10/0.0	499.	6704.	1.48
	200	<b>69</b> /2.5/0.0	493.	3294.	0.99
	50	<b>34.2</b> /0.0/0.0	245.	1720.	0.68

TABLE III. For the same plasma conditions and gas mixtures of table ??, mean ion charge of plasma, argon and added gas, and calculated percentages of argon and added gas. Top table is for optimized extracted  $\text{Ar}^{14+}$  ion currents, mid-table  $\text{Ar}^{11+}$  and bottom table  $\text{Ar}^{8+}$  ion currents.

gas	$P_{HF}$ (W)	$q_{eff}$	$q_{Ar}$	$q_{ag}$	$\alpha_{Ar}$ (%)	$\alpha_{ag}$ (%)
(i)	800	8.48	8.51	3.05	98.5	1.5
	600	8.46	8.48	3.02	98.5	1.5
(ii)	800	9.53	10.48	3.65	68.3	31.7
	600	9.06	10.29	4.01	61.5	38.5
	400	7.39	9.31	4.36	42.5	57.5
	200	7.35	8.84	3.71	50.6	49.4
(iii)	800	8.94	10.77	4.40	50.3	49.7
	600	8.66	10.46	4.47	49.8	50.2
	400	7.28	9.39	4.11	39.7	60.3
(iv)	800	8.02	10.44	5.00	37.4	62.6
(i)	800	8.47	8.50	3.04	98.5	1.5
	600	8.50	8.52	2.96	99.0	1.0
	400	8.06	8.08	3.09	99.0	1.0
	200	7.00	7.02	2.78	99.0	1.0
(ii)	800	8.96	9.43	3.80	81.5	18.5
	600	9.32	9.98	3.71	77.3	22.7
	400	8.35	9.31	4.00	66.0	34.0
	200	7.80	8.59	3.42	68.8	31.2
(i)	800	7.10	7.13	3.04	98.3	1.7
	600	6.84	6.87	3.18	98.3	1.7
	400	7.90	7.91	3.11	99.5	0.5
	200	7.03	7.04	2.91	99.4	0.6
	50	5.89	5.91	2.21	98.6	1.4
(ii)	800	8.80	9.00	3.38	91.0	9.0
	600	8.54	8.73	3.61	91.5	8.5
	400	8.08	8.22	3.47	93.3	6.7
	200	7.30	7.46	3.10	91.6	8.4
	50	6.52	6.98	2.95	76.6	23.4
(iii)	800	6.97	7.24	3.49	86.1	13.9
	600	6.92	7.20	3.46	85.6	14.4
	400	7.02	7.35	3.53	83.5	16.5
	200	6.26	6.52	3.02	85.2	14.8
	50	5.99	6.10	2.67	93.0	7.0

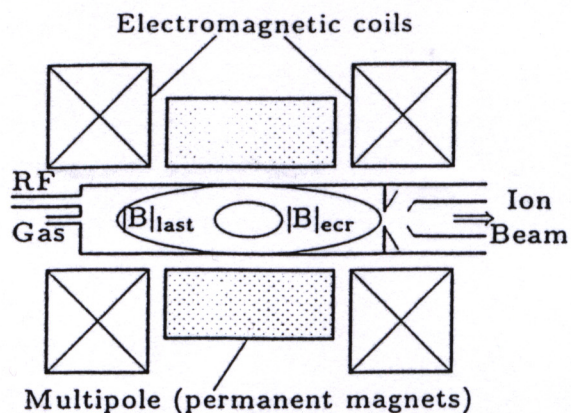


FIG. 1. Sketch of an ECR ion source :  $|B|_{last}$  refers to the highest closed  $|B|$  surface of electron confinement.

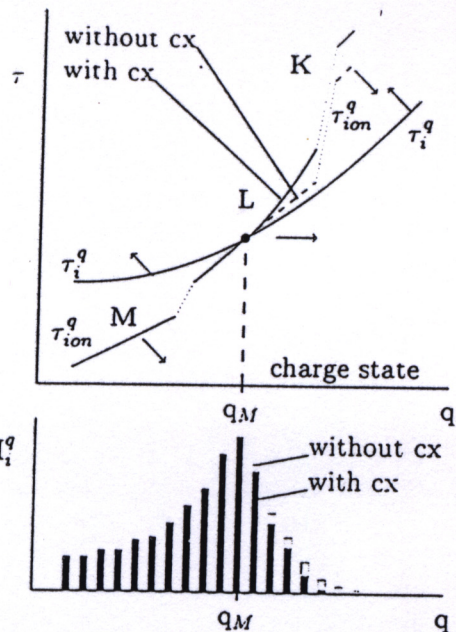


FIG. 2. Diagram showing how to determine the charge state  $q_M$  of the most abundant ion current extracted from ECRIS, from the crossover of ionization and ion confinement times. The arrows indicate how to change these times in order to increase  $q_M$ . The effect of charge exchange losses (cx) is indicated.

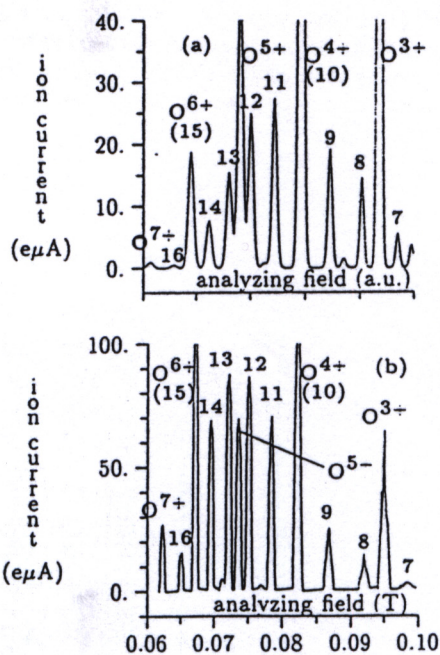


FIG. 3. Argon ion current distributions delivered by (a) Caprice source, (b) SERSE source, while optimizing  $Ar^{16+}$  ion current with  $^{16}O$  as a mixing gas.

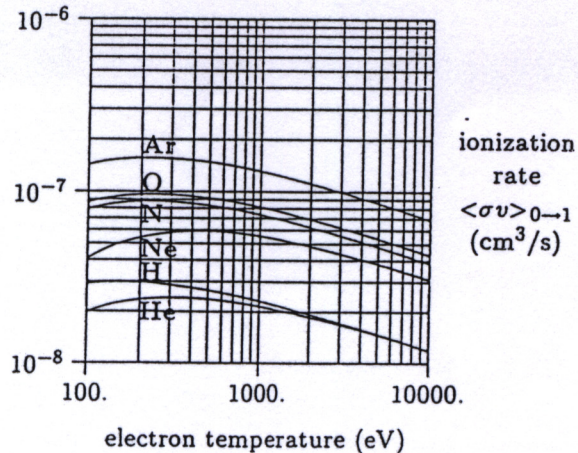


FIG. 4. First ionization rates of a few gases



TABLE III. For the same plasma conditions and gas mixtures of table II, mean ion charge of plasma, argon and added gas, and calculated percentages of argon and added gas. Top table is for optimized extracted  $\text{Ar}^{14+}$  ion currents, mid-table  $\text{Ar}^{11+}$  and bottom table  $\text{Ar}^{8+}$  ion currents.

gas	$P_{HF}$ (W)	$q_{eff}$	$q_{Ar}$	$q_{ag}$	$\alpha_{Ar}$ (%)	$\alpha_{ag}$ (%)
(i)	800	8.48	8.51	3.05	98.5	1.5
	600	8.46	8.48	3.02	98.5	1.5
(ii)	800	9.53	10.48	3.65	68.3	31.7
	600	9.06	10.29	4.01	61.5	38.5
	400	7.39	9.31	4.36	42.5	57.5
	200	7.35	8.84	3.71	50.6	49.4
(iii)	800	8.94	10.77	4.40	50.3	49.7
	600	8.66	10.46	4.47	49.8	50.2
	400	7.28	9.39	4.11	39.7	60.3
(iv)	800	8.02	10.44	5.00	37.4	62.6
(i)	800	8.47	8.50	3.04	98.5	1.5
	600	8.50	8.52	2.96	99.0	1.0
	400	8.06	8.08	3.09	99.0	1.0
	200	7.00	7.02	2.78	99.0	1.0
(ii)	800	8.96	9.43	3.80	81.5	18.5
	600	9.32	9.98	3.71	77.3	22.7
	400	8.35	9.31	4.00	66.0	34.0
	200	7.80	8.59	3.42	68.8	31.2
(i)	800	7.10	7.13	3.04	98.3	1.7
	600	6.84	6.87	3.18	98.3	1.7
	400	7.90	7.91	3.11	99.5	0.5
	200	7.03	7.04	2.91	99.4	0.6
	50	5.89	5.91	2.21	98.6	1.4
(ii)	800	8.80	9.00	3.38	91.0	9.0
	600	8.54	8.73	3.61	91.5	8.5
	400	8.08	8.22	3.47	93.3	6.7
	200	7.30	7.46	3.10	91.6	8.4
	50	6.52	6.98	2.95	76.6	23.4
(iii)	800	6.97	7.24	3.49	86.1	13.9
	600	6.92	7.20	3.46	85.6	14.4
	400	7.02	7.35	3.53	83.5	16.5
	200	6.26	6.52	3.02	85.2	14.8
	50	5.99	6.10	2.67	93.0	7.0

TABLE I. Typical  $\text{Ar}^{10+}$  ion parameters.

$\nu_{ei}(\text{s}^{-1})$	$\nu_{eqei}(\text{s}^{-1})$	$f_{ci}(\text{s}^{-1})$	$\rho_i(\text{mm})$	$\Delta l(\text{mm})$
$43 \cdot 10^6$	0.28	$1.5 \cdot 10^6$	0.16	0.04

TABLE II. Data of gas mixing experiments : plasma conditions, gas mixtures (i) pure Ar, (ii) Ar +  $^{16}\text{O}$ , (iii) Ar +  $^{18}\text{O}$ , (iv) Ar +  $^{22}\text{Ne}$ , and input rf power settings. Measured extracted Ar ion currents in (e $\mu\text{A}$ ) when optimizing only one charge state (that of bold numbers).  $\text{Ar}^{14+}$  (top table),  $\text{Ar}^{11+}$  (mid-table),  $\text{Ar}^{8+}$  (bottom table). Calculated parameters : total particle loss rate  $\sum I_i^q/q$  in (p $\mu\text{A}$ ), expression (ion energy from electrons)  $P_{ei} = \sum_i \sum_q I_i^q q^3/A_i$ , ion temperature  $T_i$  from formula (13).

gas	$P_{HF}$ (W)	$I_{Ar}^q(\text{e}\mu\text{A})$ 8/11/14	$\sum I_i^q/q$ (p $\mu\text{A}$ )	$P_{ei}$ (a.u.)	$T_i$ (eV)
(i)	800	132/31/0.68	265.	8134.	2.29
	600	145/32/0.66	246.	8615.	2.19
(ii)	800	43.6/47/5.6	353.	8477.	2.13
	600	49/46.6/4.2	366.	8846.	1.95
	400	57/23/0.60	415.	7513.	1.58
	200	53/13/0.23	364.	4609.	1.18
(iii)	800	23/35/7.0	340.	7691.	2.07
	600	32/35/5.0	335.	7929.	1.93
	400	32/15/0.80	365.	4683.	1.43
(iv)	800	19/20.4/2.6	320.	5736.	1.93
(i)	800	130/31/0.60	264.	8116.	2.29
	600	144/33/0.70	246.	8755.	2.20
	400	182/20/0.15	262.	8514.	1.91
	200	110/2.4/0.0	285.	4358.	1.26
(ii)	800	116/54/2.2	355.	10890.	2.28
	600	76/56/4.0	328.	10058.	2.10
	400	93/37/1.0	361.	8817.	1.75
	200	82/15/0.14	339.	5294.	1.26
(i)	800	181/5.2/0.0	405.	7285.	1.97
	600	165/2.5/0.0	414.	6797.	1.77
	400	187/14/0.08	267.	8015.	1.87
	200	112/2.5/0.0	284.	4435.	1.27
	50	28/0.03/0.0	212.	1606.	0.70
(ii)	800	160/51.3/1.1	321.	11390.	2.39
	600	160/38/0.7	326.	10166.	2.12
	400	200/20/0.1	304.	9387.	1.88
	200	136/5.4/0.03	319.	5399.	1.30
	50	51/0.0/0.0	292.	2291.	0.70
(iii)	800	182/6.6/0.0	498.	7130.	1.84
	600	190/9.3/0.0	515.	7629.	1.71
	400	160/10/0.0	499.	6704.	1.48
	200	69/2.5/0.0	493.	3294.	0.99
	50	34.2/0.0/0.0	245.	1720.	0.68